A Hybrid Physics-Based, Data-Driven Approach to Model Damage Accumulation in Corrosion of Polymeric Adhesives

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MICHIGAN STATE UNIVERSITY

BOSCH

Endurica
Get Durability Right

2020 U.S. DOE Vehicle Technologies Office
Annual Merit Review

Project ID #: MAT152 June 3, 2020

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Project overview

Partners

- Michigan State University (Lead)
- Robert Bosch LLC.
- Endurica LLC.
- JdV Lightweight Strategies, LLC.
- Composite Center at MSU

Budget

Total Project Funding: $1,442,188

- DOE Share: $967,662
- Collaborators Share: $474,526
- Cost Share: 32.9%
- FY 2020 DOE Share: $612,311

Timeline

Start: January 2019
End: December 2021
Completion: 41%

Barriers*

1. Lack of reliable joining technology for dissimilar materials
2. Lack of cost-effective tests for evaluation of corrosion
3. Lack of constitutive model capable of predicting corrosion
4. Predictive modeling tools
    - Prediction error <10%
    - Lack of validated test protocols

Read *Light Duty Workshop Final Report and U.S. DRIVE MTT Roadmap, section 5.1.

Relevance & Objectives

Overall Objectives:

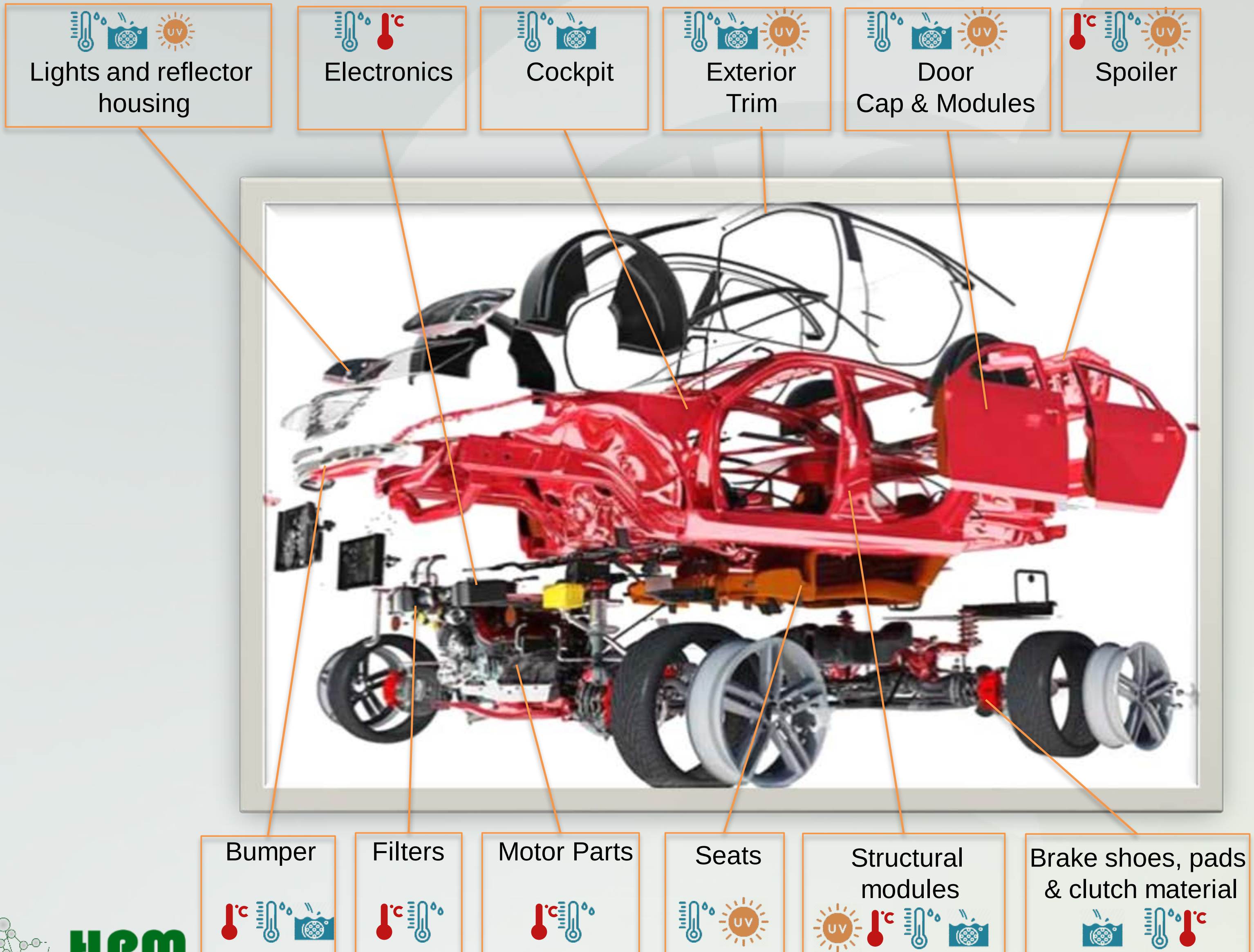
- ❖ A software to predict corrosion-induced failure in cross-linked polymeric adhesives with respect to damage accumulation by corrosion and fatigue with a 10% error.
- ❖ A theoretical model to describe damage accumulation in constitutive behavior with respect to (1) deformation, (2) vibration, (3) hydrolysis, (4) thermo-oxidation and (5) photo-oxidation.

Impact/Relevance to DOE

Predicting corrosion failure in joints of dissimilar materials is necessary to

- facilitate use of lightweight material for vehicle mass reduction
- Speed up the application of composites in vehicle structures for lightweighting to address DOE 2030 targets
- reduce time required for testing corrosion failure which makes the use of lightweight materials more attractive for OEM
- Improve CAE prediction capability to achieve a reliable design of joints

Critical segments



Aging Mech	
Thermo	°C
Hydro	
Hgyro	
Photo	UV

Approach & Milestones

In Progress	+	blue
Finished	✓	green

FY-2019		FY-2020		FY-2021		
Development	Individual Damage Mechanism	Platform Assembly	Recursive Neural Network	Characteriz-ation Failure	Framework Assembly	UMAT software design
Development of a theoretical platform for constitutive modeling of adhesives ✓	Quasi-Static deformation ✓ Vibration + Thermo-oxidative ✓ Photo-oxidative + Hydrolysis + BOSCH	Hydro+ Thermo + Photo+ Thermo + Photo + Thermo + Hydro + BOSCH	Predict corrosion-induced failure with multi-vector input parameters BOSCH	1. Effect of environment al factor + 2. Effect of different cyclic loading ✓ 3. Virtual Database + +	Validation of hybrid platform on combined mechanism in the lab Endurica BOSCH	Software predictions under lab condition against sample adhesives exposed to all combination mechanisms Endurica
FY-2019	FY-2020	FY-2020	FY-2021	FY-2021		FY-2021

Micro- to macro- scale modeling, Central Experimental Database

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Milestones

Completed	
In-progess	
Planned	

Finished FY19	Derivation & Validation of the quasi-static model	Milestone
	Derivation & Validation of the vibration induced damage model	Milestone
	Derivation of the Hydrolysis model	Milestone
	Derivation & Validation of Thermo-oxidation model with multiple adhesives	Go/No-Go
Ongoing FY20	Validation of Hydrolysis model with multiple adhesives	Go/No-Go
	Validation of the modular platform concept(finished)	Milestone
	Accumulative Damage Failure Model	Milestone
	Derivation & Validation of photo-oxidation model with multiple adhesives	Go/No-Go
	Validation of Fatigue Failure model on samples with no degradation	Milestone
	Derivation of coupled Thermo- & photo-oxidative model	Milestone
	Derivation of coupled Thermo-oxidative & Hydrolysis model	Milestone
Planned FY21	Training/Fitting Neural network engine on samples with different degradation	
	Validation of hybrid platform on combined degradation mechanisms, lab and outdoor	
	Software predictions against sample adhesives exposed to all combination mechanisms for all degradation mechanisms	

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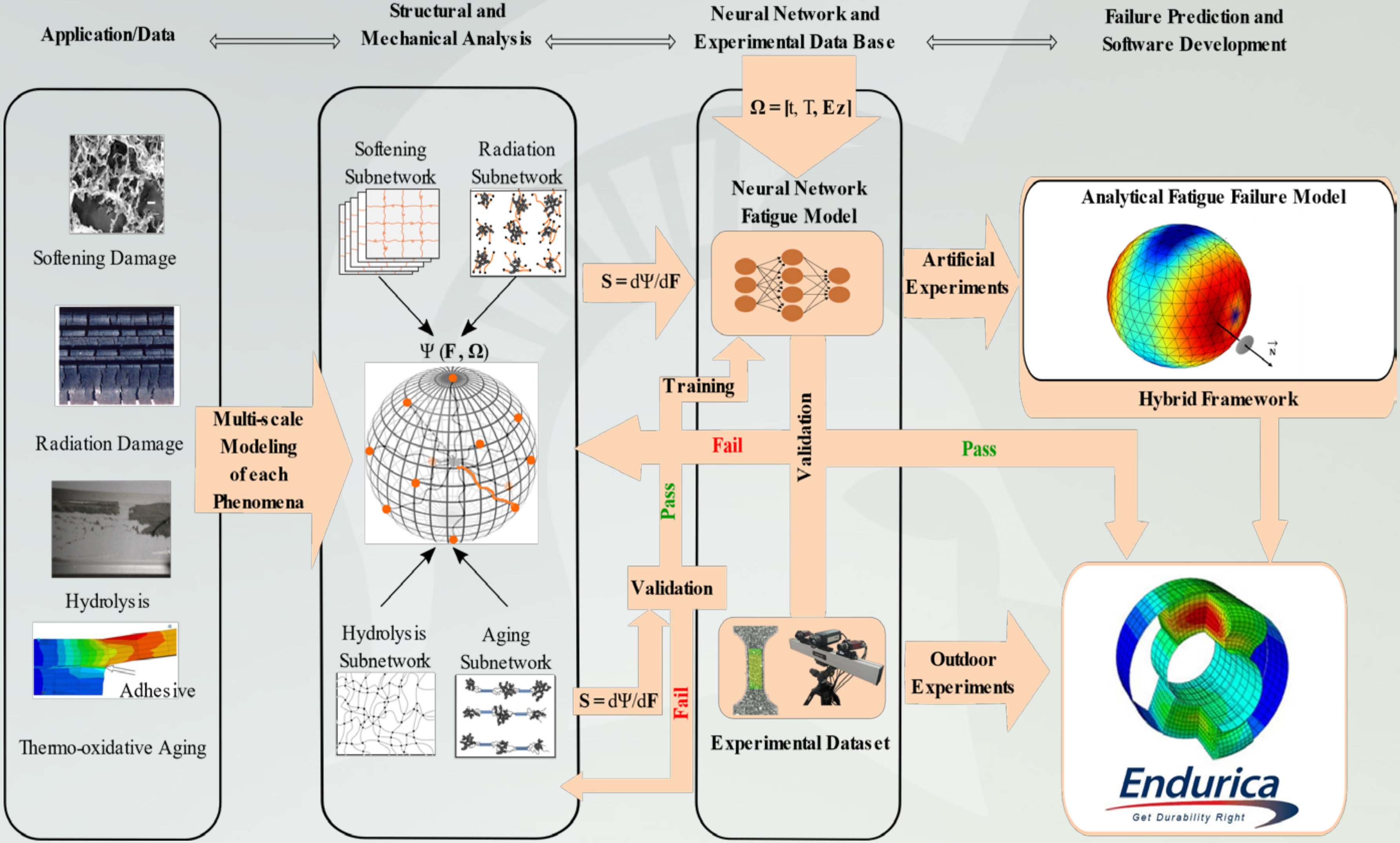
Any proposed future work is subject to change based on funding levels.

Approach- Modeling



Approach - Experimental

Adhesive pool

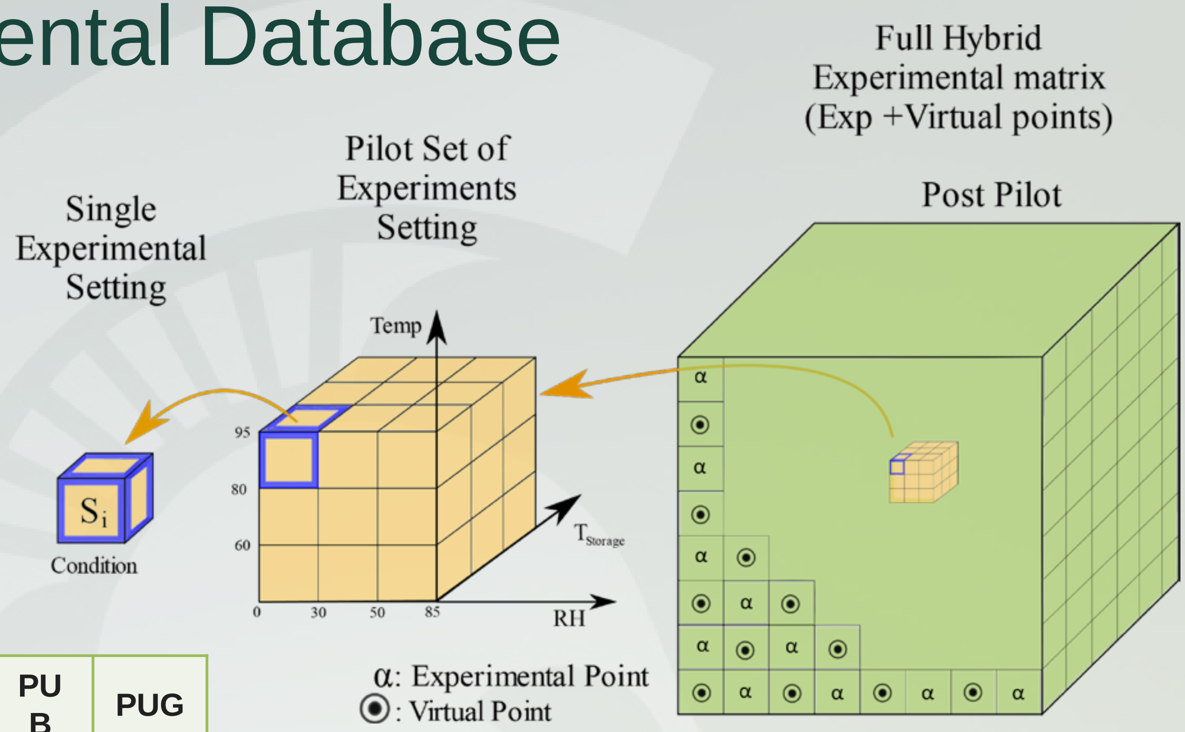
Company	Product number	type
LORD	810	Acrylic (ACR)
Dow Corning	DOWSILTM 7091	Silicon (DC)
3M	DP 6310NS	Urethane (PUG)
3M	590	Urethane (PUB)
LORD	Versilok 253/254	Acrylic
LORD	Versilok 271/331	Acrylic
LORD	850	Acrylic
LORD	320/322	Epoxy
LORD	310-A/310-B	Epoxy
LORD	320/310-B	Epoxy
3M	550	Urethane
3M	560	Urethane

Selection Criteria

	PI	Consult-ant	Collabo-rators	Industry
Application		✓	✓	✓
Manufacturer Recommend.			✓	✓
Damage resolution	✓		✓	
Reproducab-ility of results	✓		✓	✓

Central Experimental Database

Condition	Range
$T_{storage}$	1 day – 2 years
Relative Humidity	0 – 80 %
Temperature	-5 – 200 C
UV	1 – 2 kW/m^2/nm



Test Type \ Material		ACR	DC	PUB	PUG
Mechanical Tests	Reliability Test	✓	✓	✓	✓
	Failure Test for Virgin Material	✓	✓	✓	✓
	Failure Test for Aged Material	✓	✓	✓	✓
	Cyclic Test	✓	✓	✓	✓

Test Type \ Material		ACR	DC	PUB	PUG
Chemical tests	FTIR	30%	30%	30%	30%
	DSC	10%	10%	10%	10%
	Cross link Density Measurement	10%	10%	10%	10%

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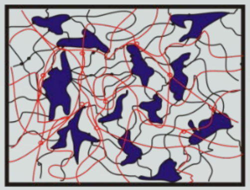
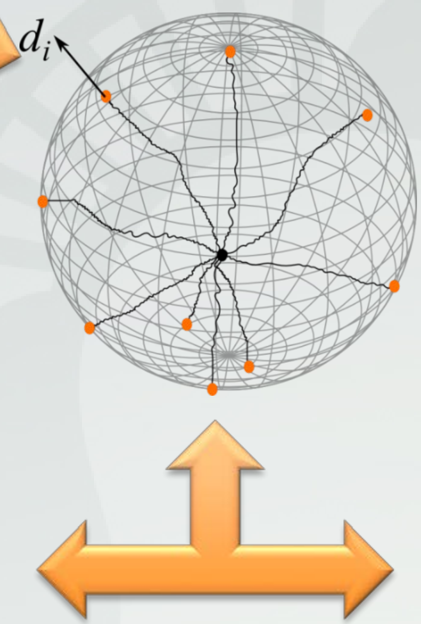
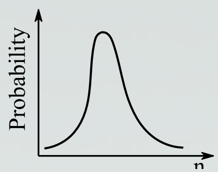
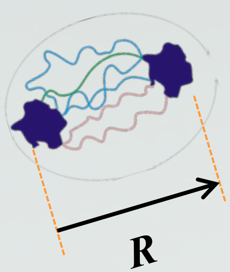
3D- to 1D- scale transition



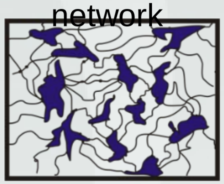


$$\Psi_M = \frac{1}{A_S}\int_S W_M^{\vec{d}}\, du^{\vec{d}} \cong \sum_{i=1}^{k} W_M^{\vec{d}_i}\, \omega_i$$

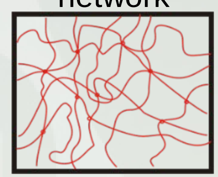


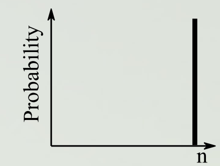


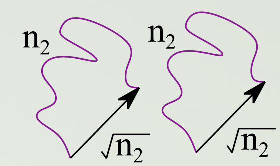
PP network



CC network







Chains distribution among two aggregates

$$W_M^{\vec{d}_i} = \int_{D_A(\lambda_{max}^{\vec{d}_i})} N(n)\psi_c(n,\bar{r})dn$$

Strain energy of a chain with n segments and end to-end distance $\bar{r}$

Chains distribution

Set of all possible chain lengths

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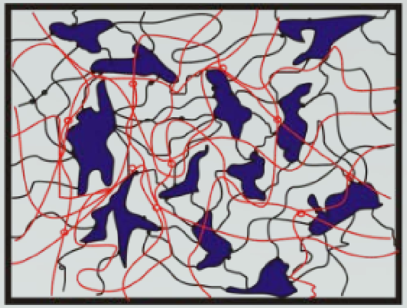
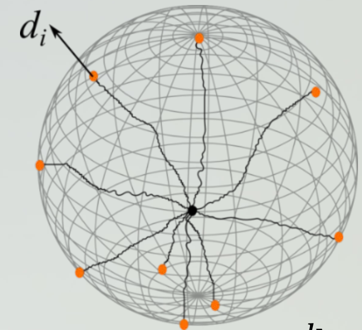
Concept and validation

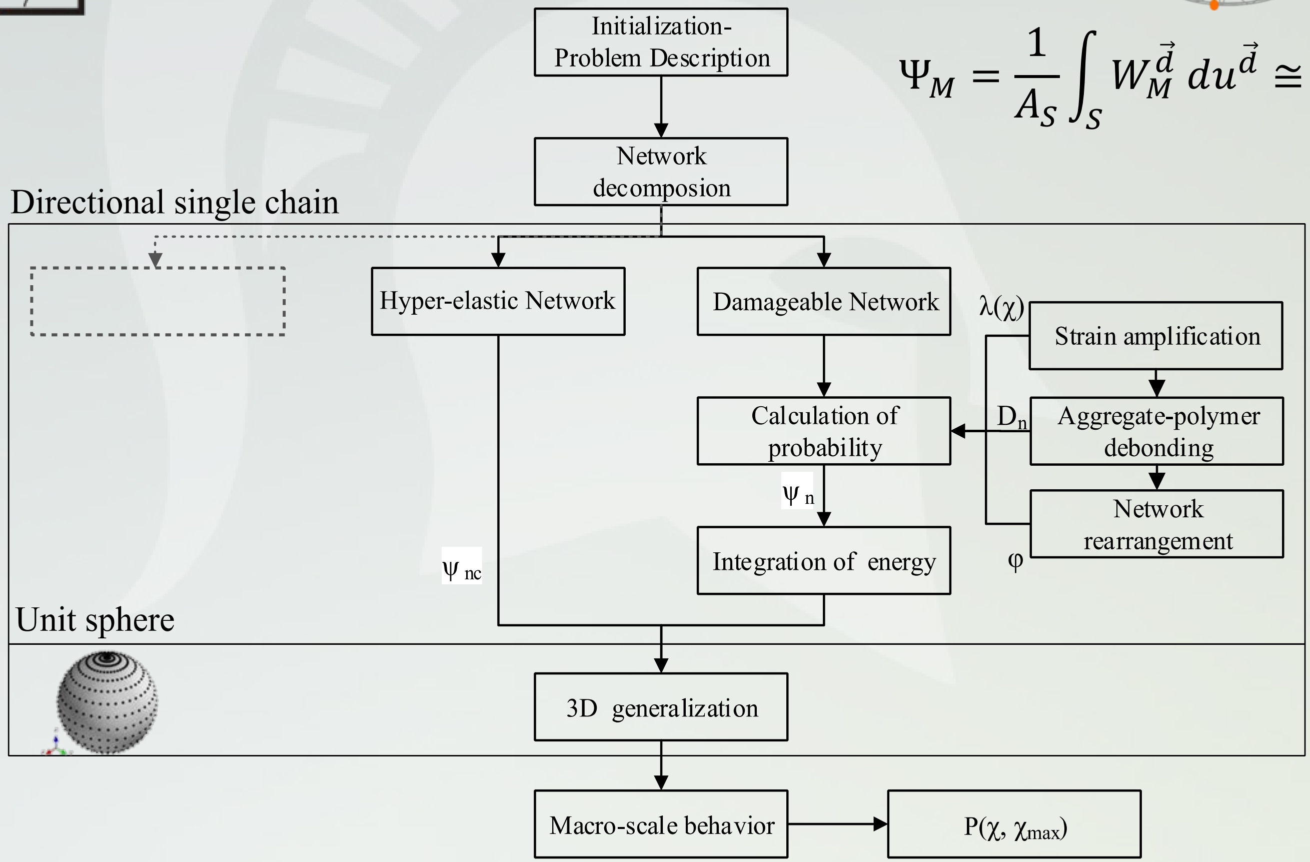


$$\Psi_M = \Psi_1 + \Psi_2 + \dots$$

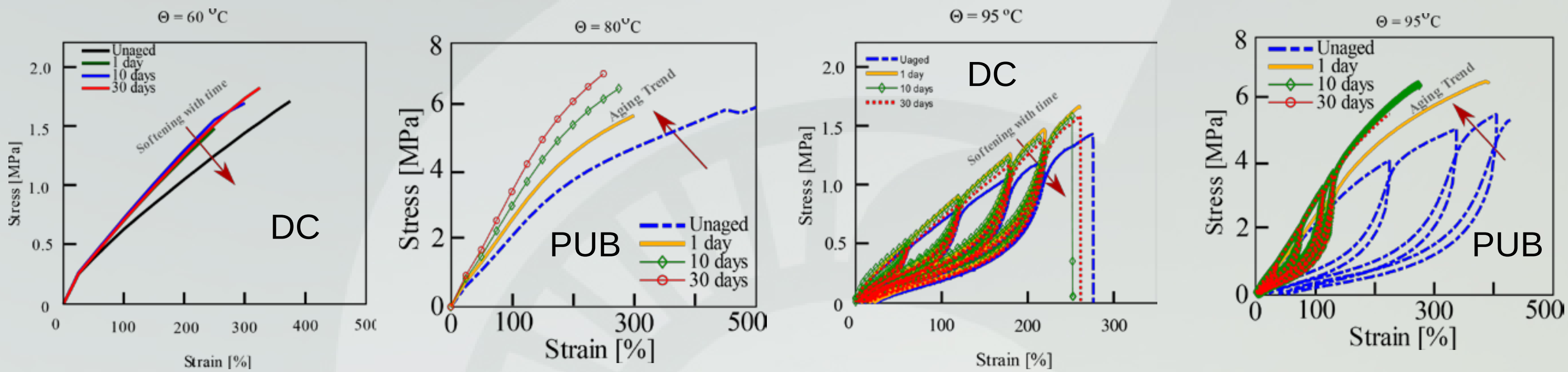
...



$$\Psi_M = \frac{1}{A_S}\int_S W_M^{\vec{d}}\, du^{\vec{d}} \cong \sum_{i=1}^{k} W_M^{\vec{d}_i}\, \omega_i$$



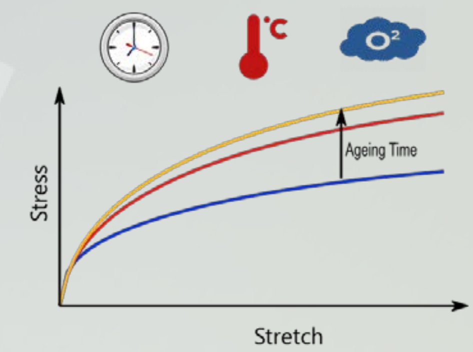
Thermo-oxidative Experiments



Observations for mechanical tests:

- Continuous hardening occurs for PUB
- Initial overcuring followed by softening occurs for DC
- Initial Loss in ultimate strength and strain for DC
- Increase in ultimate strength, accompanied by drop in ultimate strength for PUB
- Deterioration in the material exhibited as softening can be observed at higher temperatures for both materials

Thermo-oxidation Model

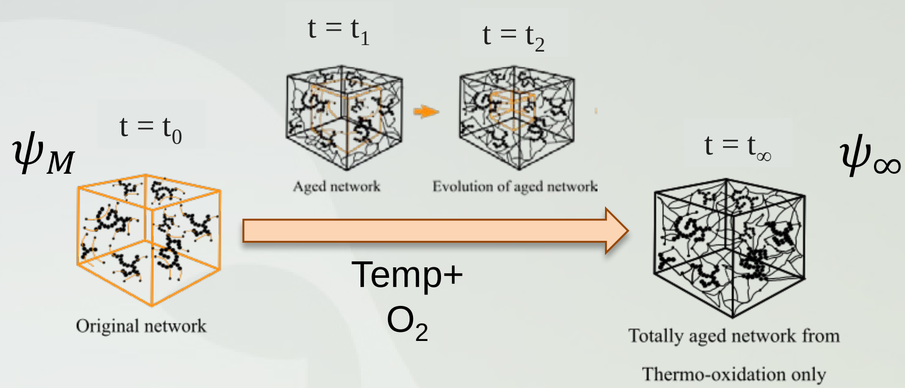


Symptoms

- Embrittlement
- Network rearrangement
- Polymer scission/reformation

Challenges

- Accelerated aging tests are not reliable
- Diffusion limited oxidation should be excluded
- Chemical anomalies must be ruled out
- Lack of global decay function



Dual network hypothesis

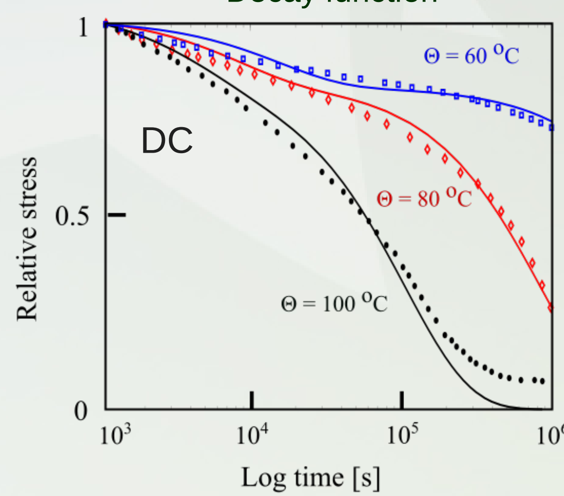
- $\psi_M = \rho_0(t)\,\psi_0 + \rho_\infty(t)\,\psi_\infty$
- ψ_M is the whole strain energy of the matrix
- $\rho_{\blacksquare}$ is the concentration of each network

ψ_0

- $-\frac{d[p]}{dt} = k[p]^n$
- $[p]$ is the concentration
- of chemical Factors
- $k = \tau_0 \, exp\left(-\frac{E_a}{RT}\right)$

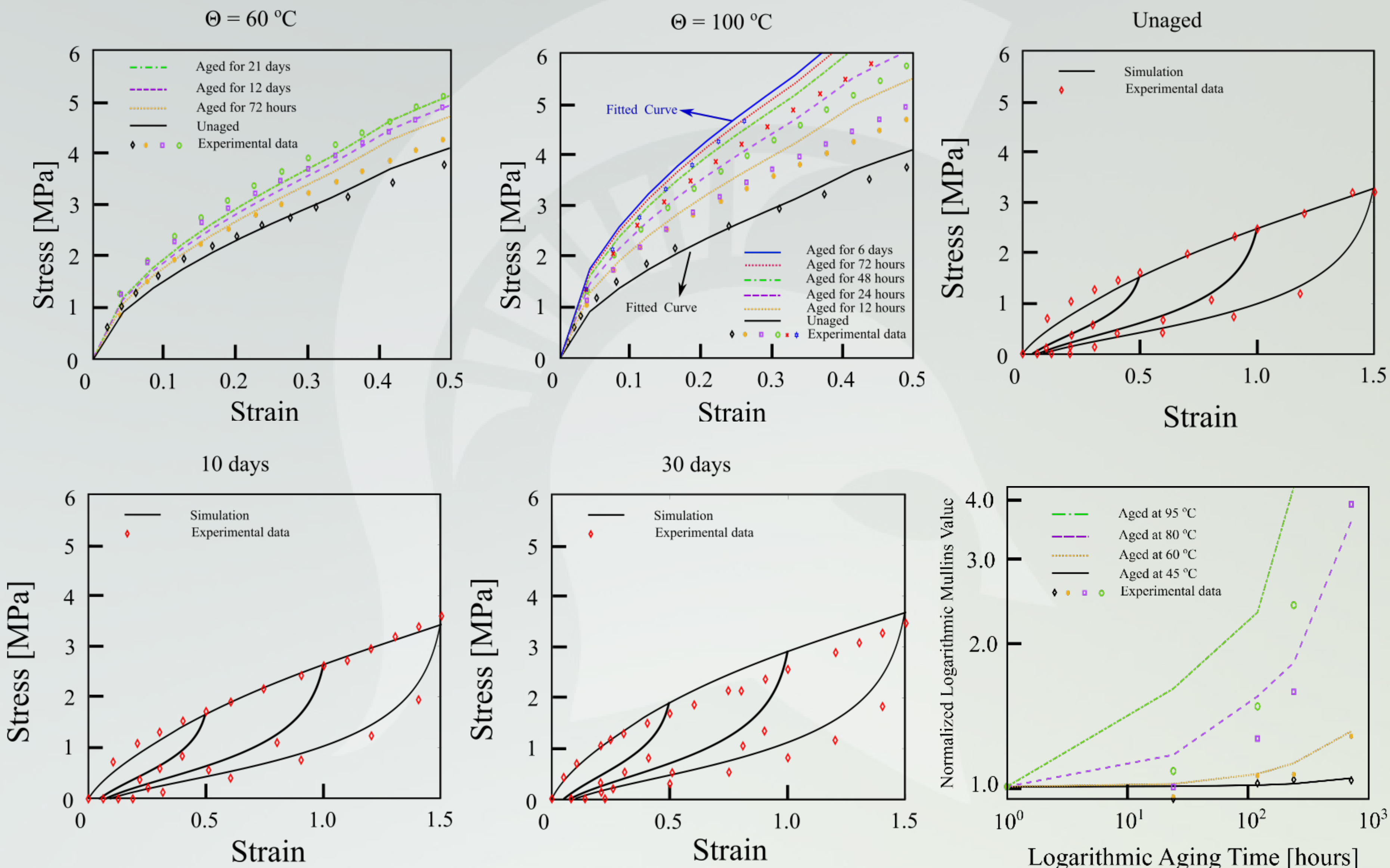
E_a: activation energy
R : is the ideal gas constant
τ_0: is pre-exponential factor
n : is the order of reaction

Decay function



Average error at 60
2%
Average error at 80
9%
Average error at 100
37%

Thermo-Oxidation Model Validation- Intermittent Tests



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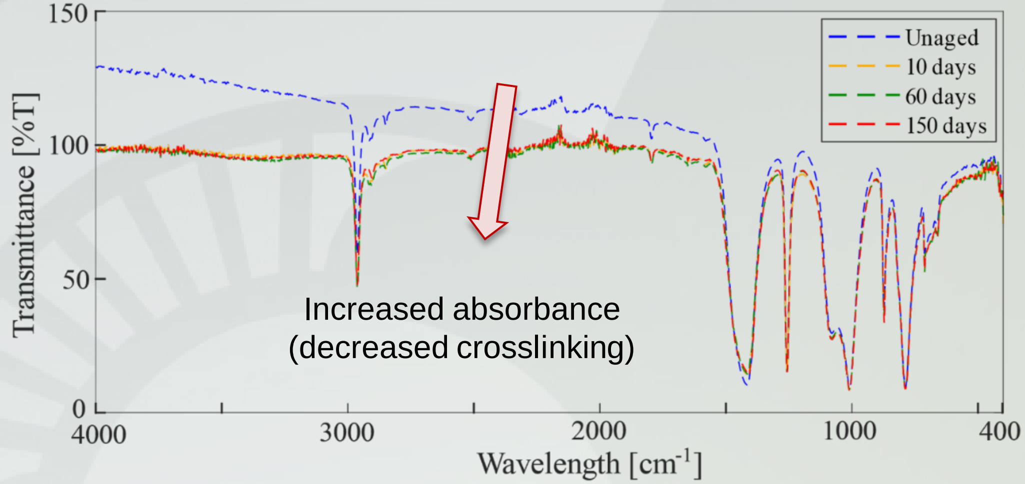
Plots of stress change due to aging (Modeling vs experimental Results)

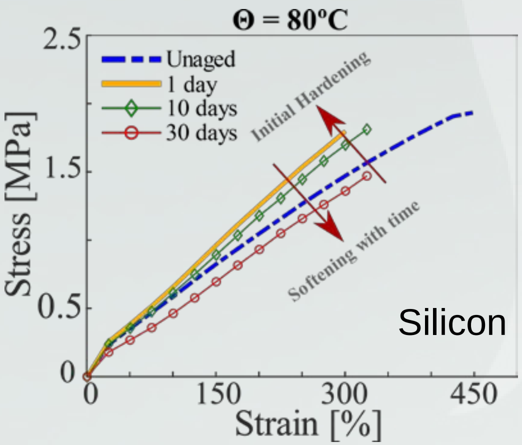
Photo-oxidation Experiments

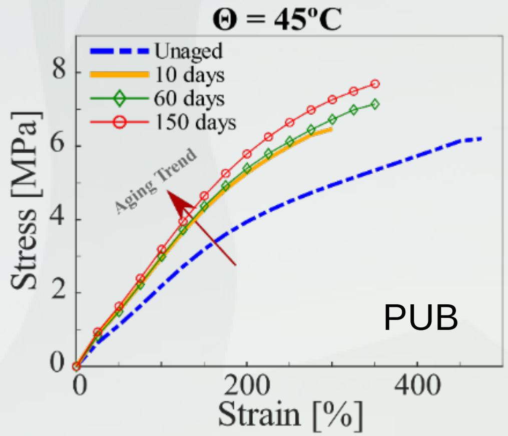
Chemical Tests
(FTIR test for DC)

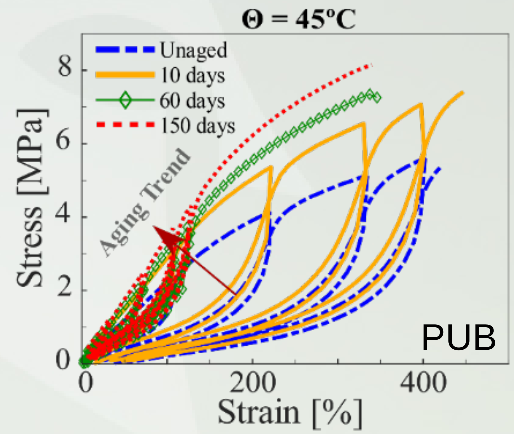
UV Machine



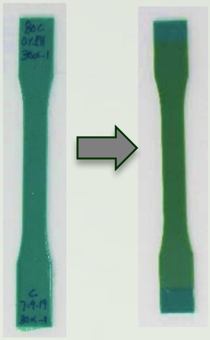
Mechanical Tests







Sample Discoloration



Observations:

- Over-curing results in material hardening
- Photo/oxidative damage can cause hardening or softening
- Higher temperatures speed up the damage process

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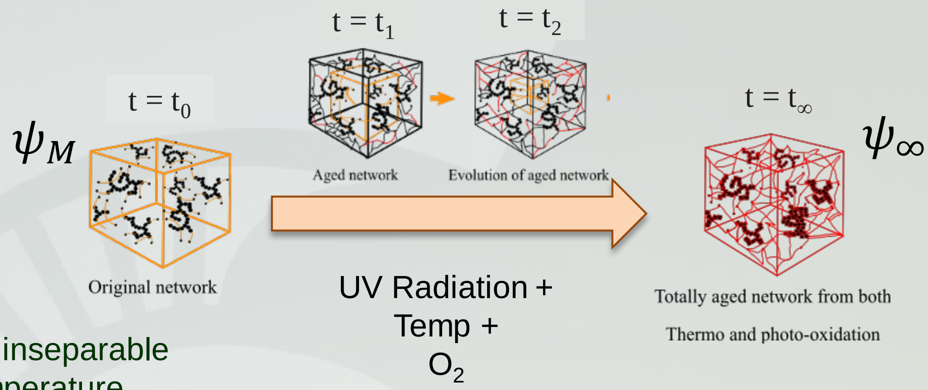
Photo-oxidative Model

- Losing properties through time
- Chain scission
- Decrease of cross-link density



❖ Challenges

- Effect of thermo- and photo-oxidative is inseparable
- Mechanism of aging dependency to temperature
- Lack of experimental data
- Inconsistency in experimental results

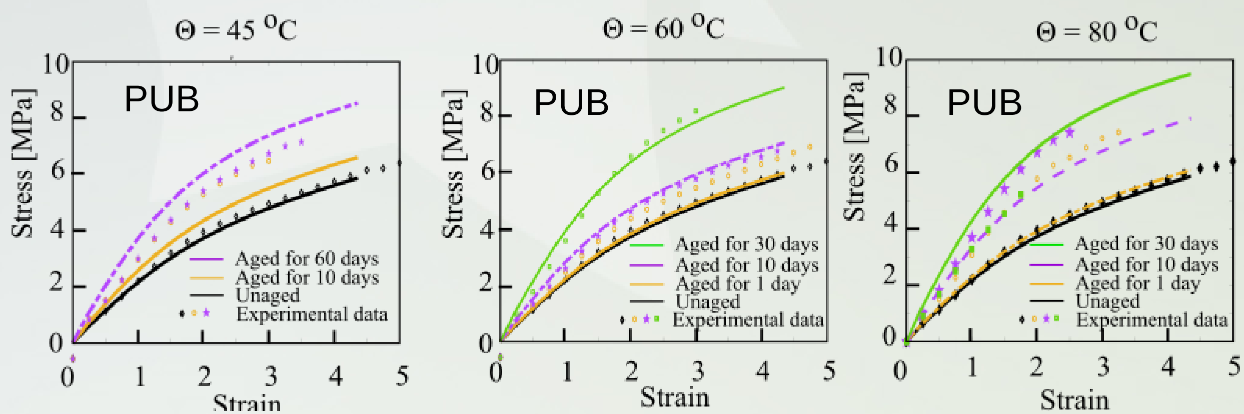
❖ Concept and validation

$$\varphi_{photo+thermo} = \rho_{thermo}\varphi_0 + (1-\rho_{thermo})\big(\rho_{photo}\varphi_{thermo} + (1-\rho_{photo})\varphi_{photo}\big)$$

$$\rho_{thermo} = A_1 \exp(-e^{\frac{-E_a}{RT}} t)$$

$$\rho_{photo} = A_2 \exp(-I^{\alpha} t)$$

- A_1, A_2, α : Constants
- I : Radiation intensity
- E_a : Activation energy



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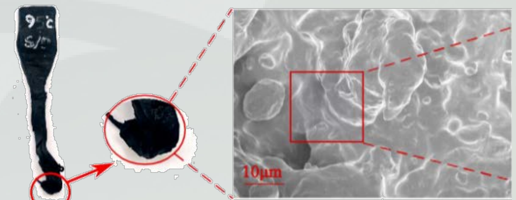
Hydrolysis Experiments

Damage in the polymer matrix will take place with respect to two different mechanisms;

i) Deformation-induced damage

ii) Environmental-induced damage



(d) (e)

Relative stress softening σ^*

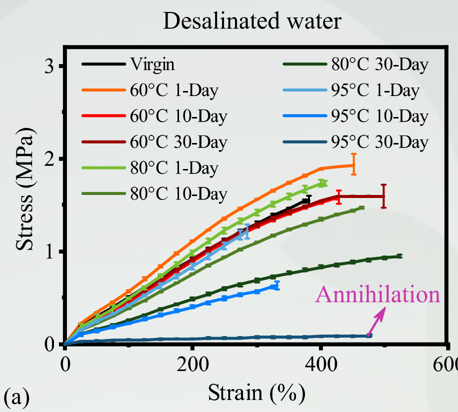
$$\sigma^* = \frac{\sigma_1}{\sigma_{max}}$$

Relative Residual strain e^*

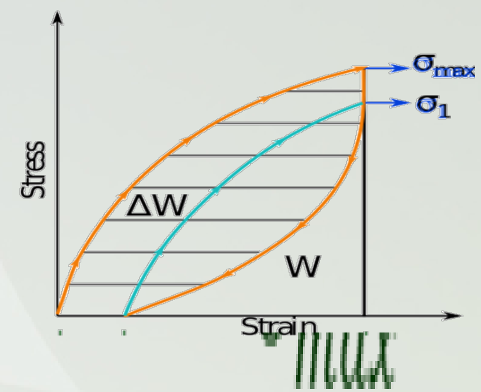
$$e^* = \frac{e_1}{e_{max}}$$

Hysteresis Loss W*

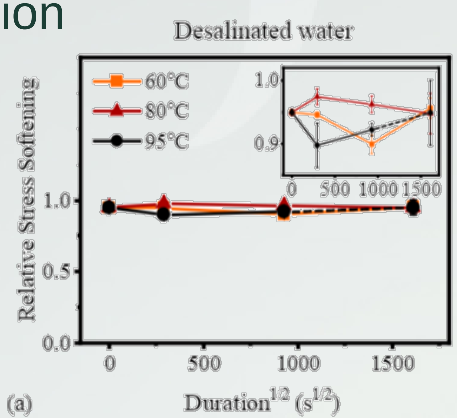
$$W^* = \frac{W}{\Delta W}$$



(a)



❖ Validation



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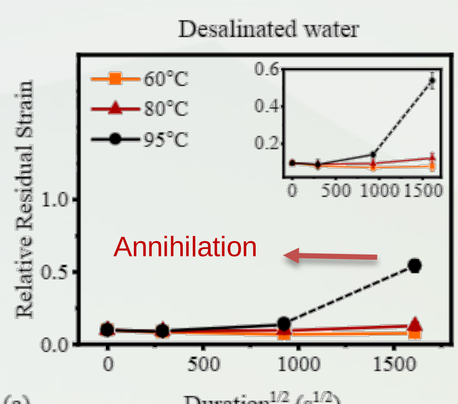
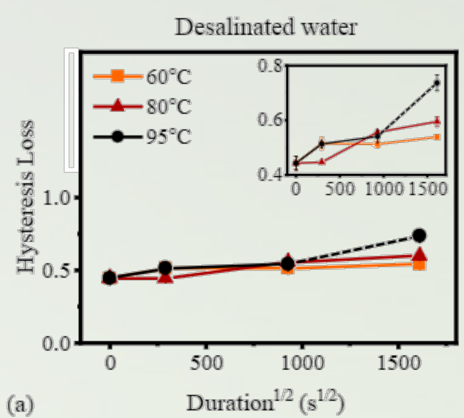


(a)



(a)

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Hydrolysis Model

$$\Psi_M(t,T,\mathbf{F}) = N(t,T)\Psi_0(\mathbf{F}) + N'(t,T)\Psi_\infty(\mathbf{F})$$

$$N(t,T) = \exp\left(-\gamma \exp(-\frac{E_a}{\mathcal{R}T})t\right)$$

- Strain energy of a single chain

$$\hat{\psi}_c(n,\bar{r}_\bullet) = nK_bT\int_0^{\varphi}\hat{\beta}d\varphi, \quad \hat{\beta} = \left[1 - \frac{1+\varphi^2}{n}\right]\beta$$

- Probability Distribution Function of a Polymer Chain

$$\mathcal{P}_\bullet(n) = \frac{1}{2\sqrt{\pi\sigma^2}}\exp(\frac{(n-\mu_\bullet)^2}{-2\sigma^2})$$

- Networks and Subnetworks

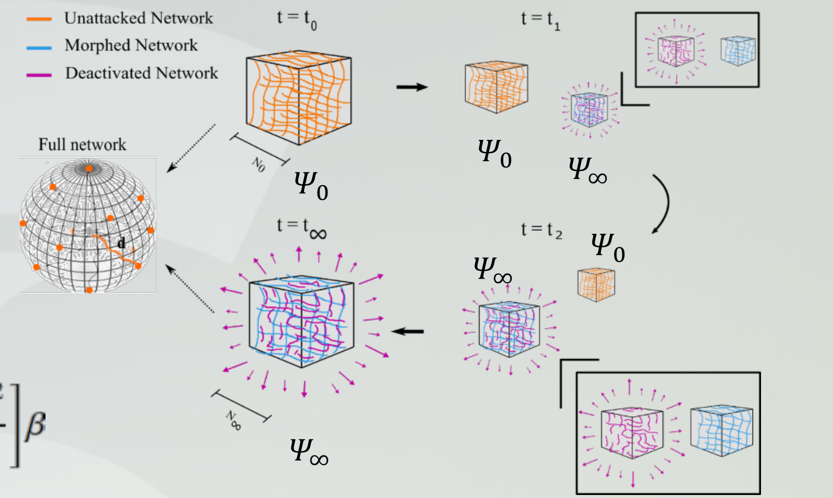
$$\Psi_\bullet = \frac{1}{A_s}\int_S \overset{d}{\psi}_\bullet \overset{d}{du} \cong \sum_{i=1}^{k}\overset{d_i}{\psi}_\bullet w_i$$

- Inverse Langevinge Function approximation

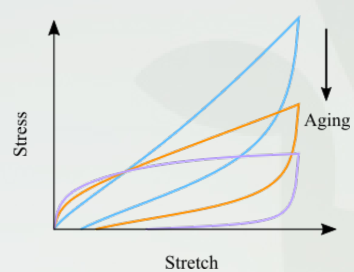
$$\mathcal{L}^{-1}(x) \cong \frac{1}{1-x} + x - \frac{8}{9}x^2$$

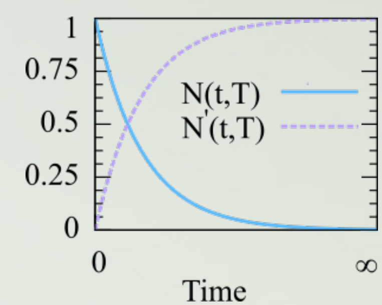
- Kintetics (Esters, Amide, Imide,Carbonate)

$$-\frac{d[COOH]}{dt} = \xi[Ester][Water][COOH] = \kappa[COOH]$$



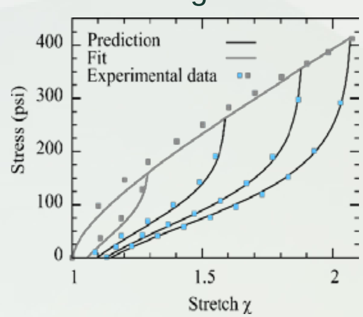
$$\Psi_M = N(t,T)\Psi_0 + \alpha\, N'(t,T)\Psi_m + \ (1-\alpha)N'(t,T)\Psi_d$$



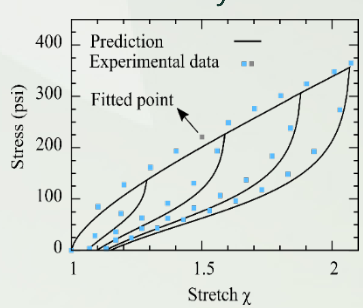


Θ= 60 ° C

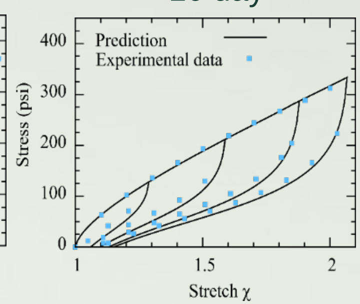
Unaged



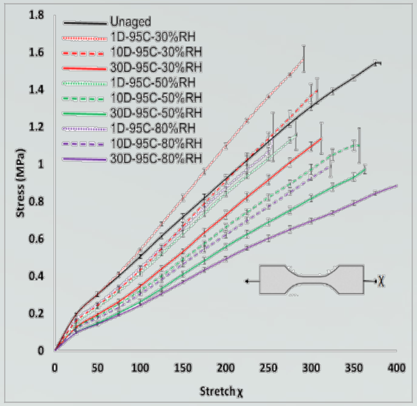
6 days

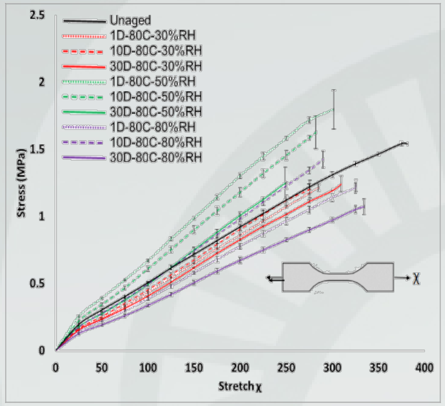


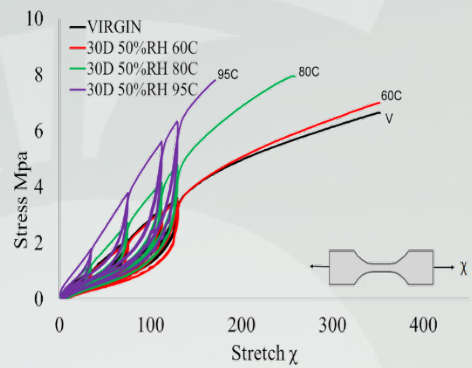
10 day

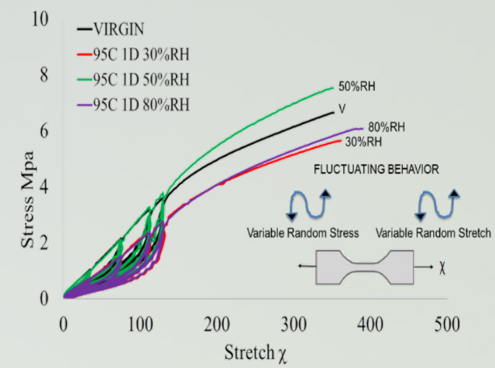


Hygrothermal Experiments









Observations for Silicon adhesive:

- Initial over-curing
- Softening with time
- Loss in ultimate strength
- Loss in ultimate strain

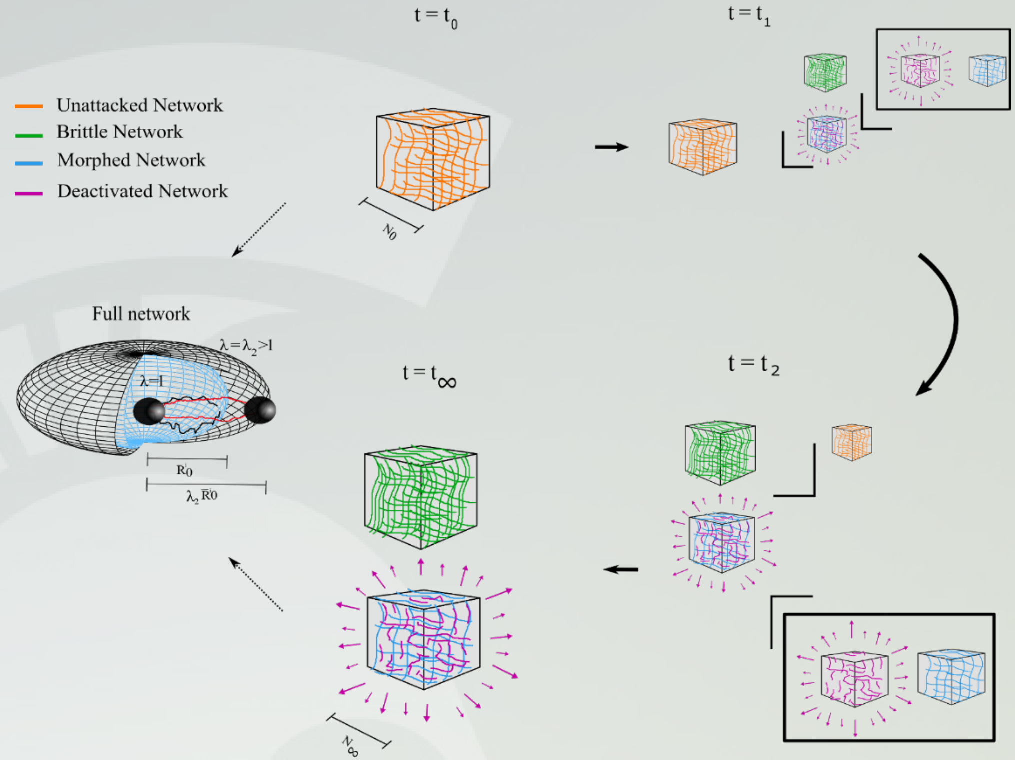
Observations Polyurethane adhesive

- Higher temperature results in increased hardening with time
- Higher temp result in loss in ultimate stretch

Hygrothermal Model

"Accelerated aging using moisture and heat cycles"

- loss of the mechanical performance
- of polymeric materials.
- Reduction of cross-link density
- Chain scission due to oxygen attack on backbone
- Increased rate of degradation due to oxidation



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$$N(t,T) = \exp\left(-\gamma \exp(-\frac{E_a}{RT})t\right)$$

$$\Psi_\infty = (1-\beta)\Psi_T + \beta\alpha\Psi_m$$

$$\beta = \theta RH\sqrt{t}\exp\left(\frac{-E_b}{RT}\right)$$

Zero Humidity Submerged condition

$$0 < \beta < 1$$

$$\Psi_M(t,T,\boldsymbol{F}) = \exp\left(-\gamma\, exp\left(\frac{E_a}{RT}\right)t\right)\Psi_0 + \left(1-\exp\left(-\gamma\, exp\left(\frac{E_a}{RT}\right)t\right)\right)\left\{\left(\left(1-\theta RH\sqrt{t}\exp\left(\frac{-E_b}{RT}\right)\right)\Psi_T + \theta RH\sqrt{t}\exp\left(\frac{-E_b}{RT}\right)\alpha\Psi_m\right)\right\}$$

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Central Exp. Dataset- Virtual datapoints

Experiment design

L Experiment conditions

S1t_1 D1 T1 RH1 UV1
t1 D2 T1 RH1 UV1
t1 D1 T2 RH1 UV1
…

Experimental investigation

Di Ti RHi Uvi
Mechanical tests
Chemical Tests

Feature Extraction

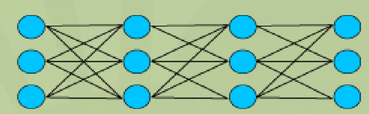
Condense the experimental data based on change on the micro structure of material

- **Tg**
- **Specific peaks in FTIR**
- **Toughness**
- **Energy dissipation**
- **Viscoelastic parameters**

⋮

Learning process

Inputs
$S_i = \{t_s, T, RH, UV, \ldots\}$

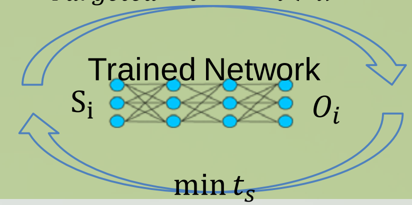


outputs
$O_i = \{\mathrm{D}, T_g, FTIR, \delta, \ldots\}$

Minimization process

Aging time minimization to yield same properties

$O_{Targeted}(S_t) = O_i(S_i)$?



$S_i \Big|_{O_t(S_t)=O_i(S_i)}$

$t) = O_i(S_i)$

Challenges of Blackbox Neural Network(NN) Engines in constitutive modeling

- Incomplete Data (Mapping n-dimensional chebyshev space into m-dimensions)
- Polyconvexity
- Frame-indifference
- Convergence

α: Experimental Point
⦿: Virtual Point

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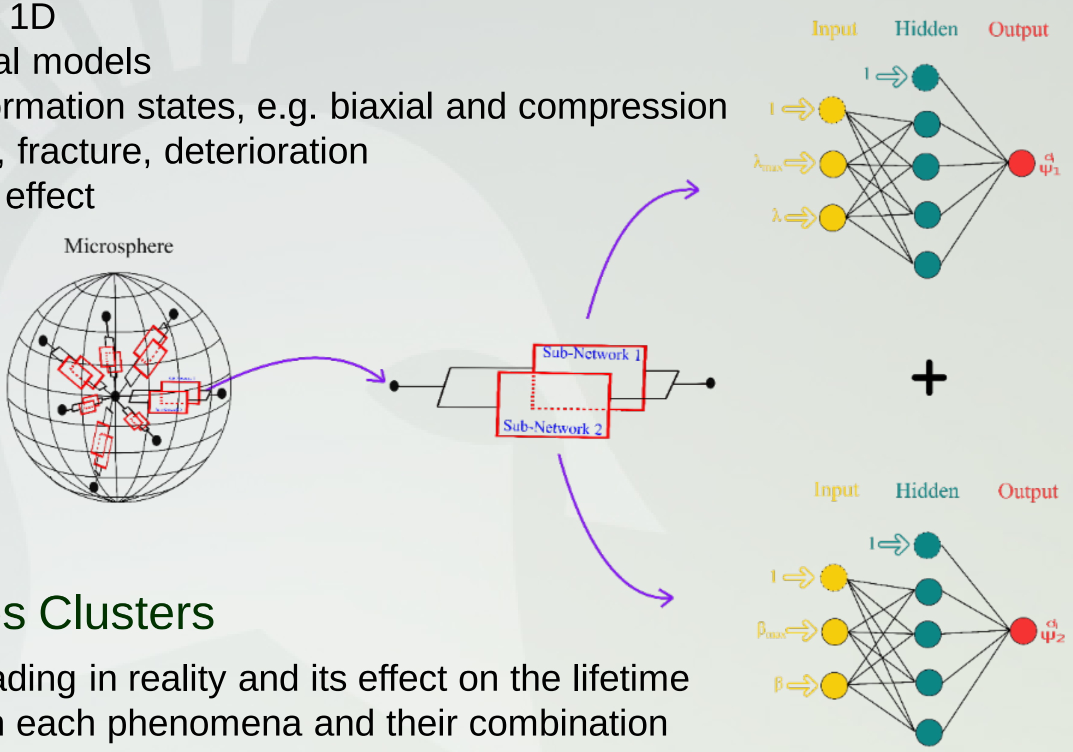
Physics-informed Neural Network Engines

Proposing a physics-informed cluster of super-simplified NN engines

- Valid for amorphous networks
- Micro sphere model convert 3D to 1D
- Simplify complex micro-mechanical models
- Two subnetwork to predict all deformation states, e.g. biaxial and compression
- Just useful for directional damage, fracture, deterioration
- Inelastic behavior such as Mullins effect



Implementation of NN engines Clusters

- Non-stationary and 3-Dloading in reality and its effect on the lifetime
- Limited number of data on each phenomena and their combination
- Crack generation due to environmental conditions
- Size-effect (the models are developed based on the assumption of uniform aging in the material)
- Complicated nature of aging with multiple agents

2020 Progress	
2019 Progress	

Technical Accomplishments

In Progress

Modular Platform publication

- Khalili et al. (2019), Rubber Chem. & Tech. 92(1), 51-68
- Morovati & Dargazany (2019), SoftwareX 100229
- Morovati et al. (2019), ', Math. Mech. Solids
- Morovati & Dargazany (2019), Phys Rev. E. 100229

Vibration

- -Moravati&Dargazany IMECE2020

Thermo

- -Mohammadi & Dargazany, Int J. Plasticity, 118 (2019)

-Mohammadi et al. ECCMR 2019
-Morovati & Dargazany, IEC 2019

Hydro

- Bahrololoumi et al., Int. J. Plasticity 1. (2020)

- Bahrololoumi & Dargazany IEC 2019

Hygro

- Wanru et al. IMECE2020
- Bahrolouloumi et al. IMECE 2020

Photo

Thermo+ Photo

Thermo+ Hydro

Thermo+ Hygro

Machine learned Engine

Model Free approaches

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Response to Previous Year Review Comments

- **Approach Clarity**: *"The breadth of the approach is impressive, but it is not clear how the progress gets integrated for a cohesive tool or set of tools." "It is recommended to include data input/output flow chart among team members in next year's presentation. "*

  - ❖ Two descriptive slides were added to illustrate the integration of different parts of the project to validate and merge the concepts toward development of the predictive software.

- **Relevance to Auto. Industry :** *"Relevancy to joining with adhesives for automotive construction is missing. ""The team is highly motivated and needs proper steering towards the actual goal." "world examples of adhesive joining used in automotive" "struggling with the selection of the adhesives" "having input from car manufacturers, National labs, or Adhesive Manufactures"*

  - ❖ The team are collaborating with two of the largest adhesive manufacturers for auto industry on compound types, Test design, and characterization procedure for hybrid aging
    - ❖ Parker-Lord chemicals Inc. (NDA signed)
    - ❖ Dow Chemicals Inc.
    - ❖ Dan Houston: Auto-Manufacturing consultant

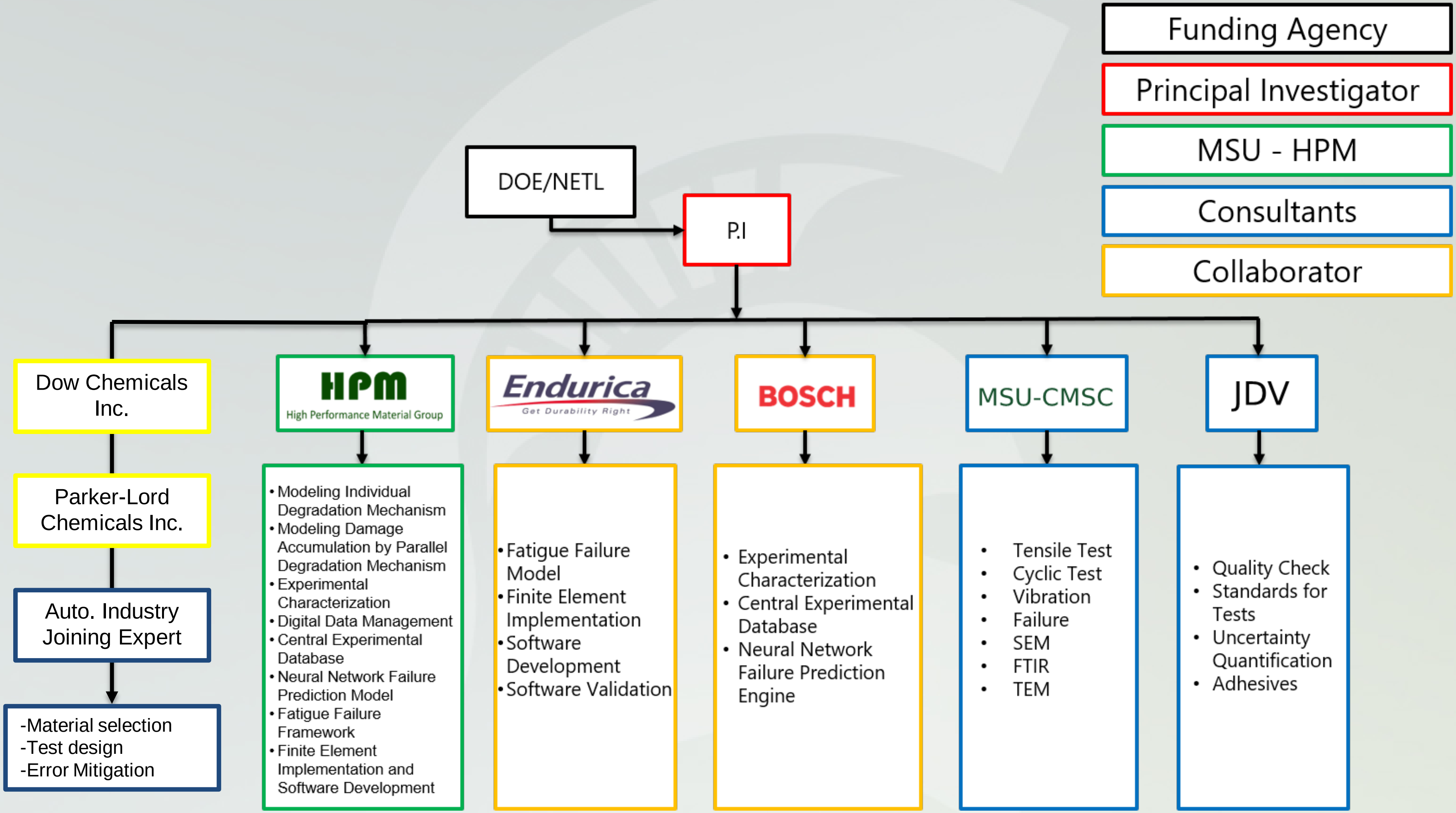
- **Cost-optimization: "***minimum critical number of experiments-> target accuracy". "it studies all failure modes of networked structured adhesive" "The project should be more focused on selecting/identifying features that can efficiently capture the underlying mechanics/physics/chemistry of the system.*

  - ❖ In pilot-phase for each compound 3,218 tests were planned. Depending on the material behaviour, sensitivity analysis will be performed to derive the number of additional necessary tests needed to train the Neural Network engine to predict the rest of the <u>Experimental Database</u>.

- **Scope limitation:** *"The project is over-ambitious". " ..whether the damage mechanism of joints/adhesives can be all related to the mechanism listed for the approach."*

  - ❖ The project is focused on understanding/simulation of bulk adhesive properties, and does not include (1) Actively varying enviormental load, (2) Interface damage, (3) delamination, (5) substrate damage.

Collaboration and Coordination



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Remaining Challenges and Barriers

COVID-19 labs shut down:

- Forced shut down of all Aging Tests
- Removal of all ultra-long aging samples
- Capacity shift by industrial collaborators (uncertainty on resource allocation)
- Budget expenditure on halted operations

Modeling	Experiment(amir and hamid and EXP)
Various nonlinear behaviors with specific features for different adhesives	The cost and complexity of corrosion mechanisms aging to achieve isolation of single mechanism
Extrapolation capabilities of the NN models	Impurities (compound, and curing) can expedite corrosion-induced failure of bulk samples
Non-uniform damage mechanism in the material	Design of hybrid accelerated tests depends on assuming similar mechanisms at different conditions
Complicated and inseparable sources of degradations mechanism	Inconsistency between accelerated and normal aging tests results

Summary

Accomplishments

- Established a systematic procedure of screening and selection of adhesives
- Finished Pilot-tests on 4 selected compounds
- Developed and validated a modular platform that allows different damage mechanisms to be integrated together
- Hypothesized/Developed & verified models of vibration and thermo-induced damage on three adhesive types
- Developed the concept development for photo-oxidation and hydrolysis damage mechanisms verified on pilot adhesive

Future Research

- Outdoor chemical, mechanical and physical characterization of bulk adhesives at different climate zones
- Development of minimized set of tests for training/validation of NN engines for different compounds
- Integration of other damage mechanisms such as bio-degradation, and diffusion limited oxidation

Any proposed future work is subject to change based on funding levels.

Technical back-up slides

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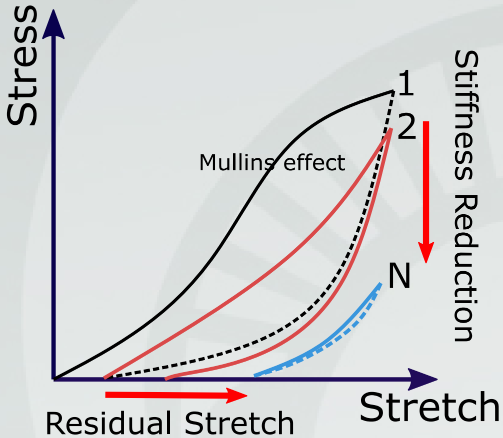
Vibration-induced damage

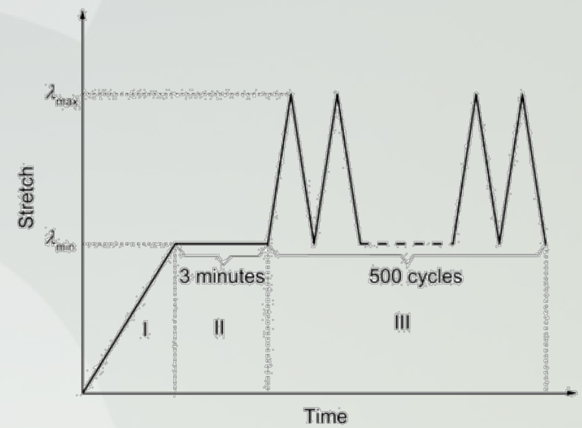
Softening of the material due to large time usage

To model the constitutive behavior of adhesives through **vibration**

Approach

Experiment :



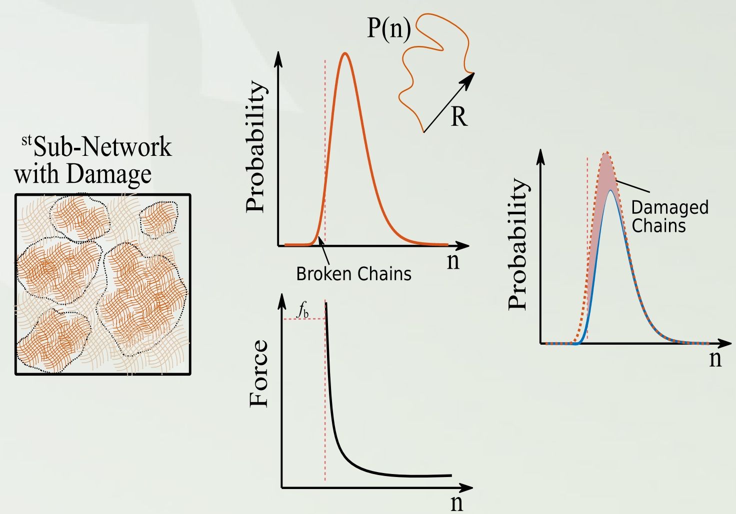


Constitutive model :

Using kinetics of irreversible chain scission

$$\overset{d_i}{\underset{\sim}{P}}(n) = P_0(n)\, e^{-C_s(n)\, j}$$

$$C_s(n) = \int_{cycle} \exp\left[\frac{\alpha}{k_B T}\left(\mathcal{L}^{-1}\left(\frac{R\lambda^{d_i}}{n}\right) - f_a\right)\right] dt$$



Thermo-Oxidative Aging

Goal: To model the constitutive behavior of adhesives through thermo-oxidative aging

Challenge

Finding the correct decay function

Approach

Dual network hypothesis

Arrhenius functions as decay function

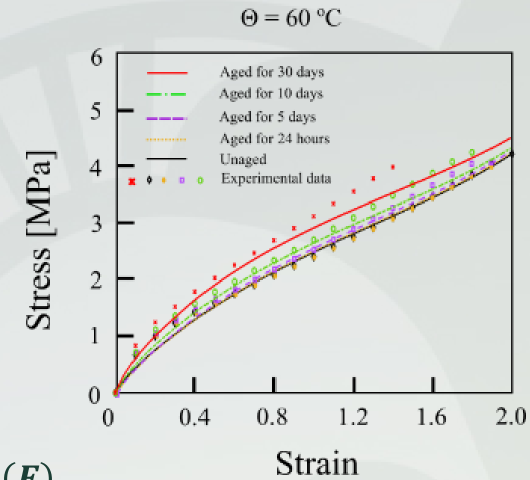
$$\Psi_M(t,T,\boldsymbol{F}) = \rho\,(t,T)\Psi_0(\boldsymbol{F}) + \big(1-\rho(t,T)\big)\Psi_\infty(\boldsymbol{F})$$

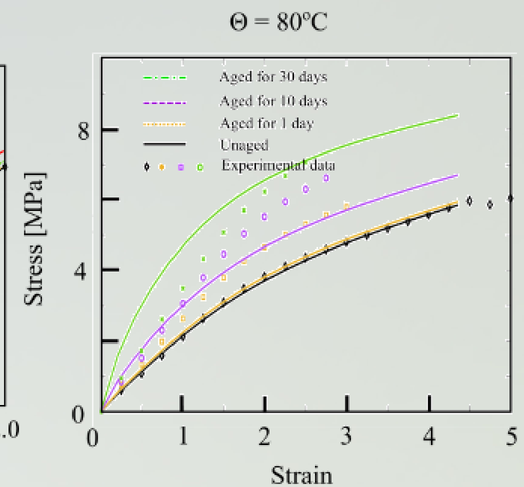
$$\rho\,(t,T) = A_1\ exp(-\alpha t) + A_2\,exp(-\beta t)$$

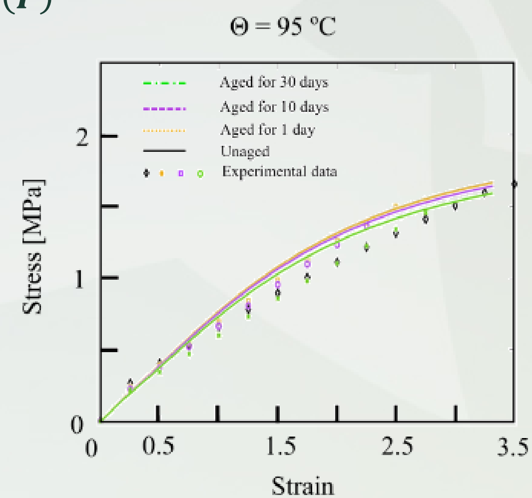
Time-temperature superposition

$$a_T = exp\left(\frac{E_a}{R}\left(\frac{1}{T_{ref}} - \frac{1}{T}\right)\right)$$

Result







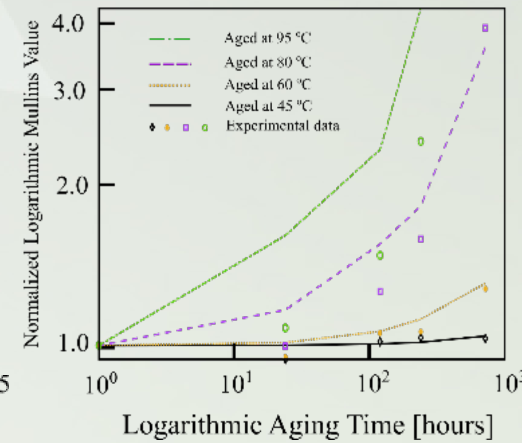


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Photo-Oxidative Aging

To model the constitutive behavior of adhesives through photo-oxidative aging

Challenge

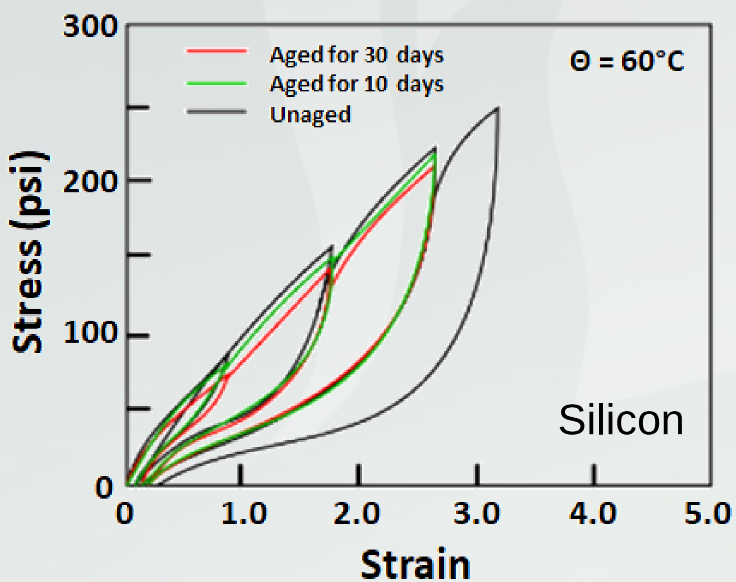
Effects of photo- and thermo-oxidation are inseparable

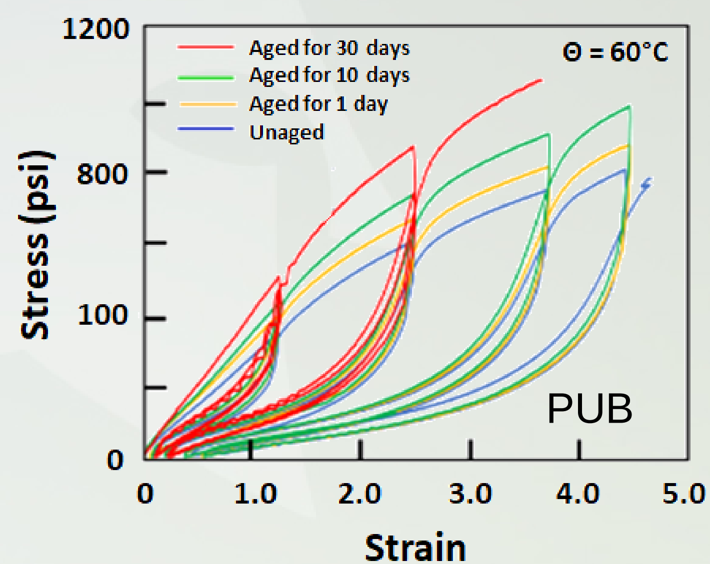
Approach

Find a decay function that can consider the effect of both phenomena

$$\rho\,(t,T) = A_1 \exp\left(-\tau_1 \exp\left(-\frac{E_{a1} + E_{photo}\,\gamma}{RT}\right)t\right) + A_2 \exp\left(-\tau_2 \exp\left(-\frac{E_{a2} + E_{photo}\,\gamma}{RT}\right)t\right)$$

Result





Constitutive behavior of the dog-bone samples

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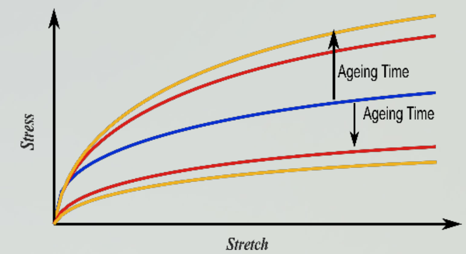
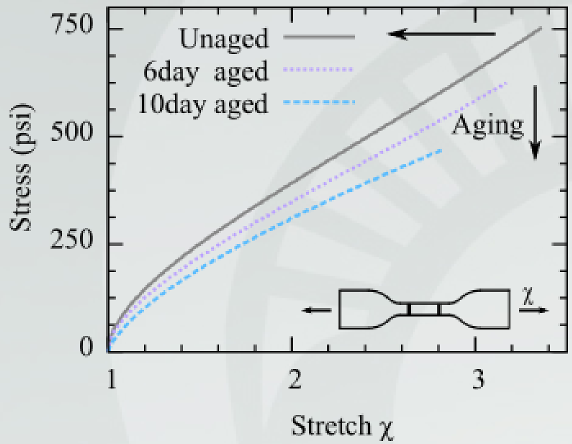
Hydrolytic Aging

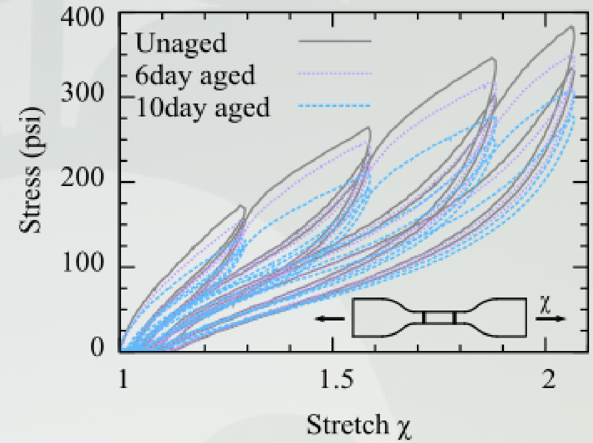


To model the constitutive behavior of adhesives through **hydrolytic** aging

Approach

Experiment :



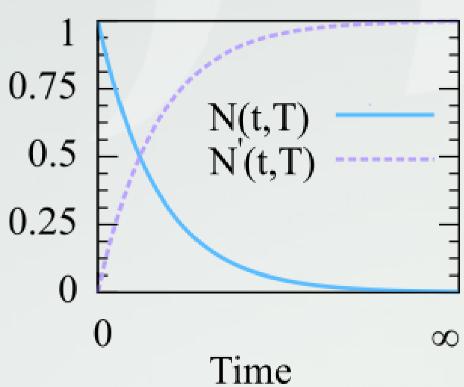


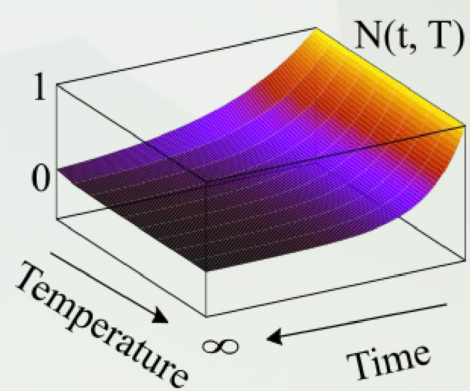
Constitutive model :

Using Arrhenius functions
as shape function

$$\Psi_{\mathrm{M}}(t,T,\boldsymbol{F}) = N(t,T)\Psi_0(\boldsymbol{F}) + N'(t,T)\Psi_\infty(\boldsymbol{F}),$$

$$\text{where} \quad N(t,T) = \exp\left(-\gamma \exp\left(-\frac{E_a}{RT}\right)t\right)$$





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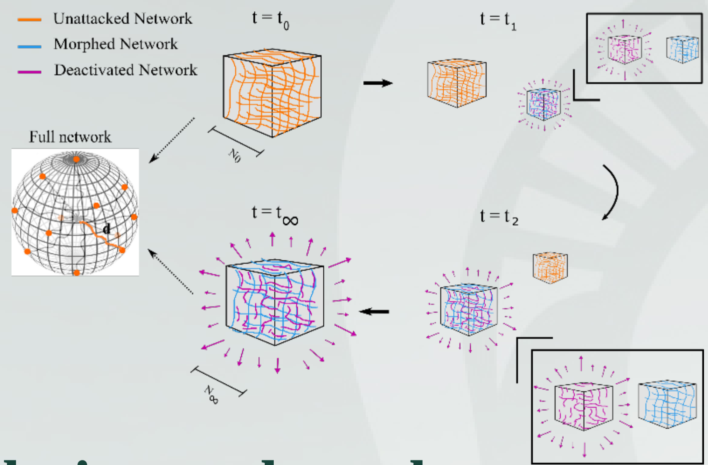
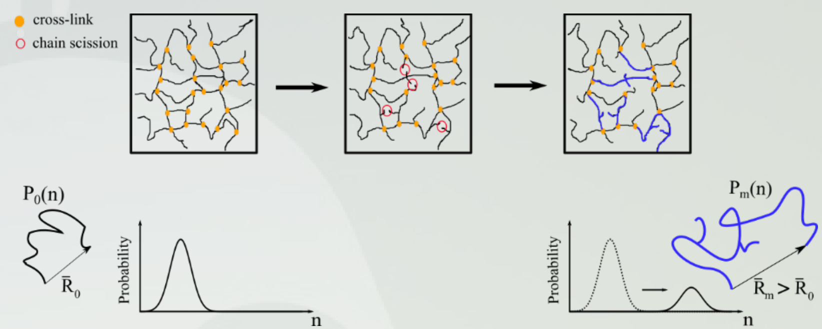
Once water attacks network, it causes the two phenomena :

(i) reduction of the cross-links, which results in a network with longer chains (morphed network)

(ii) Energy dissipation due to the reduction of active chains (deactived network)

$$\Psi_\infty = \alpha\Psi_m + (1-\alpha)\Psi_d$$





Validation and results :